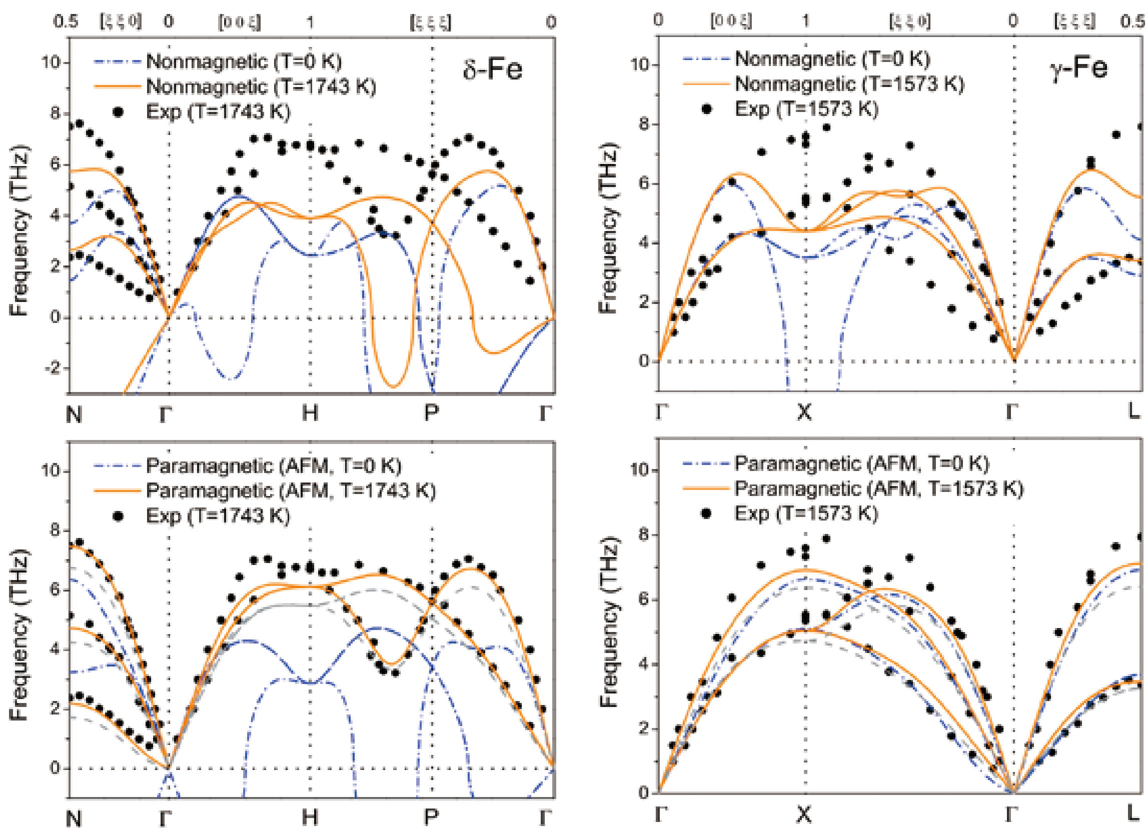
Anharmonic Lattice Dynamics of Iron at High Temperature from First-Principles

The structure and magnetism of elemental iron (Fe) undergo significant changes under high temperatures. The low-temperature α-Fe adopts a body-centered cubic (bcc) crystal lattice, and is ferromagnetic below the Curie temperature T_C = 1043 K. Upon heating, two paramagnetic structural phase transitions occur: at 1185 K to the face-centered cubic (fcc) γ-Fe and at 1667 K to the high-temperature bcc δ-Fe. Despite the ground-state behavior of α-Fe being well described by the Stoner theory of ferromagnetism, the mechanisms for the transitions to the high-temperature γ and δ phases remain unresolved. Early theoretical accounts for these phase transitions were highly controversial due to considering only magnetic or vibrational contributions to the free energy. Recent inelastic neutron

Fig.1 Calculated phonon dispersions of paramagnetic bcc δ-Fe (left) and fcc γ-Fe (right), compared to the experimental data measured at 1743 K for δ-Fe and at 1573 K for γ-Fe, respectively. The solid orange lines represent the self-consistent phonon calculations. (Image by Institute of Physics)



scattering measurements of the phonon dispersions of iron near distinct phase transitions showed that the vibrational and electronic entropies contribute almost equally to the α-γ transition, while the vibrational contribution dominates at the γ-δ transition, providing important insights into the stability mechanisms of the high-temperature iron phases. Nevertheless, the physical interactions governing these unusual thermodynamics are still poorly understood. Under the quasi-harmonic approximation, dynamical mean-field theory studies have reproduced the observed phonon behavior of iron near the α-γ transition by including the finite-temperature magnetism. The same approach failed, however, at the γ-δ transition yielding phonon dispersions differing significantly from the experimental data.

For most metals, the anharmonic lattice effect originating from phonon-phonon interactions is especially important at high temperatures, as was demonstrated in the studies of group III (Sc, Y, La) and IV (Ti, Zr, Hf) metals. Recently, Prof. WANG Jiantao's group (including graduate student LIAN Chaosheng and Prof. WANG Jiantao) in collaboration with Prof. CHEN Changfeng from the University of Nevada, Las Vegas (UNLV) performed state-of-the-art calculations of the anharmonic phonon dispersions of iron at its γ-δ phase transition using a self-consistent *ab initio* lattice dynamical method in conjunction with an effective magnetic force approach via the antiferromagnetic approximation. The calculated results show that in the high-temperature d-Fe phase, significant phonon renormalization is found due to the phonon-phonon interactions, resulting in the dynamical stability of the open bcc lattice, whereas in the close-packed fcc γ-Fe phase, only small anharmonic lattice effects are predicted, and the magnetic interactions play a more prominent role (Fig. 1). In addition, the comparisons with results under nonmagnetic approximation reveal the interplay between the magnetism of the electrons and the lattice degrees of freedom, demonstrating the previously missed interplay is responsible for the earlier failure of describing the high-temperature lattice dynamics in paramagnetic iron via considering the degrees of freedom separately. For both δ-Fe and γ-Fe, with simultaneous considerations of the lattice anharmonic and magnetic interactions, their calculated temperature-dependent lattice dynamical properties agree well with the experimental data. This work highlights the key role of lattice anharmonicity in determining the structural stability of iron at high temperatures, and will have significant implications for further studies of other high-temperature paramagnetic metals like Ce and Pu.

This study entitled "*Ab initio* study of anharmonic lattice dyanmics of iron at the γ-δ phase transition" was published on *Physical Review B* 92, 184110 (2015).

This study was supported by the National Natural Science Foundation of China (Grant No. 11274356) and the Strategic Priority Research Program of the Chinese Academy of Sciences (Grant No. XDB07000000). C.F.C. was supported in part by the U.S. Department of Energy under the Cooperative Agreement No. DE-NA0001982. The researchers are thankful to the crew of the Quantum Computational Center of the Institute of Physics, Chinese Academy of Sciences, for their support at the Shuguang supercomputing facilities.